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Attestation

6

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The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr. Patent application No. Demande de brevet n°

99306900.4

PRIORITY DOCUMENT

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**Blatt 2 der Bescheinigung
Sheet 2 of the certificate
Page 2 de l'attestation**

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See for original title of the application page 1 of the description



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TS 0793 EPC
Sb

FLOW CONTROL SYSTEM

The present invention relates to a process for controlling the flow rate of an oxidant/fuel mixture applicable in catalytic oxidation processes.

Catalytic oxidation processes such as catalytic partial oxidation are processes which are highly exothermic and therefore highly susceptible to changes in the oxidant/fuel ratio which has a direct influence on the reaction temperature. Therefore, there is a need to be able to measure the flow of an oxidant/fuel mixture not only quickly but also accurately since a slight deviation from the desired ratio may shorten the life of the catalytic partial oxidation catalyst employed. This is even more important in processes which operate under conditions which include changes, in particular frequent changes in the amount of oxidation product required depending on the specific requirements of the process.

For instance, in the case of catalytic partial oxidation processes which are directed at producing hydrogen (and carbon monoxide) which hydrogen is to be used as feedstock for a fuel cell as envisaged in hydrogen-operated motor cars, there will be a need for rapid and sometimes frequent decreases and increases in the flow of hydrogen to the fuel cell depending on the requirements of the motorist. In such situations not only a quick response to changes in operation is needed but also a response which is accurate and sustainable.

Temperature is of course one of the main classical and conventional parameters when monitoring changes in reactive behaviour of chemical processes. Reaction temperatures can be measured and monitored by various

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methods known in the art. Thermocouples are well known for temperature measurement, also at relatively high temperatures. They are reliable and normally have a long service life.

5 However, in systems which are dynamic and are operating at high temperature thermocouples are not adequate as their responses are relatively slow and therefore can not cope with the rapid changes occurring in such systems which would lead to incorrect information
10 and improper (re)-adjustments.

15 It has now been found that quick and reliable information can be obtained when use is made of the changes in colour or light intensity of the acting catalyst system which manifest themselves when the temperature of a catalytic system is influenced because of changes imposed thereupon by factors such as the oxygen/carbon ratio of the feedstock.

20 The present invention therefore relates to a process for regulating the oxygen/carbon ratio in the feedstock for a catalytic oxidation process in which a feedstock comprising a mixture of fuel and oxidant is passed through a catalyst bed in a reactor and hydrogen-containing reactor effluent is collected, in which process the oxygen/carbon ratio in the feedstock is
25 regulated by

a) measuring the temperature upstream of the catalyst by means of a quick response device,
b) feeding the measured data of the temperature measurement to a control unit and comparing them with
30 a calibrated temperature profile, and adjusting the amount of fuel in the feedstock to substantially maintain the oxygen/carbon ratio under non-steady hydrogen production.

35 The process according to the present invention can be conveniently applied in so-called dynamic catalytic

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oxidation processes, in particular in non-stationary catalytic partial oxidation processes. In catalytic partial oxidation processes much emphasis is laid on the quality and the yield of hydrogen produced (together with carbon monoxide). Whilst the composition of the hydrocarbon feedstock will govern the quality of the hydrogen-containing flow (which is *inter alia* relevant when it is intended to use part or all of it for further processing in a fuel cell to generate electricity for mobile or stationary applications such as driving vehicles or in domestic applications such as units to generate heat and/or power), it is the demand by the motorist which, in essence, dictates the amount of hydrogen to be produced for conversion in a fuel cell to produce the required amount of electricity.

Since the demands of the motorist can and will fluctuate depending on the actual circumstances, it is important that the hydrogen input to the fuel cell can be regulated as best as possible. Since catalytic partial oxidation (to be discussed in some detail hereinafter) is an exothermic process it will be clear that even a rather small deviation from a predetermined hydrogen yield (corresponding with a certain level of feedstock conversion and therefore with the oxygen/carbon ratio of the feedstock) will have a relative large effect on the temperature at which the catalytic oxidation process is carried out and hence on the amount of hydrogen produced. Since, moreover, varying amounts of hydrogen will have to be produced depending on the actual circumstances, it is of eminent importance to be able to correlate supply and demand of hydrogen as best as possible. It is also advantageous when the regulating measures applied are not only fast but also reliable and cheap.

The regulating process according to the present invention is based on the use of a quick response

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temperature device in combination with an appropriate actuator to achieve the required oxygen/carbon ratio under non-steady conditions.

5 The process is illustrated by means of Figure 1 in which in situ calibration downstream of the catalyst bed is depicted. It will be clear that the process according to the present invention is not limited to this particular embodiment since other means of calibrating can also be applied (as will be discussed hereinafter).

10 A hydrocarbonaceous fuel passing through line 1 via actuator 16 and an oxygen containing stream passing though line 2 are mixed in line 3 and the mixture is entering reactor 4 to pass through a catalyst bed (not shown) during which passage a catalytic partial oxidation process is carried out. The hydrogen-containing reactor effluent is removed via line 5 and can be subjected to further processing (such as the separation of hydrogen and carbon monoxide in unit 6 from which hydrogen can be removed via line 7; hydrogen separated can then be used 15 as input for a fuel cell to produce electricity (not shown)).

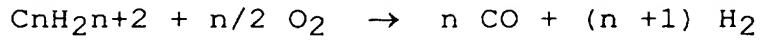
20 The regulating system as depicted in Figure 1 is based on a quick response device 10 capable of measuring the temperature upstream of the surface of the catalyst 25 in reactor 4, a means 11 to transfer data to control unit 12, a device 13 capable of measuring the temperature of the bottom of the catalyst bed, a means 14 to transfer data to control unit 12 and a transfer means 15 to adjust actuator 16 in accordance with the information processed 30 in control unit 12. It will be clear that the process is reiterative when constant O/C ratios are required and adaptive when (small) changes in the O/C ratio are required.

35 The partial oxidation of hydrocarbonaceous feedstocks, in particular hydrocarbons, in the presence of a

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catalyst is an attractive route for the preparation of mixtures of carbon monoxide and hydrogen, normally referred to as synthesis gas. The partial oxidation of paraffinic hydrocarbons is an exothermic reaction

5 represented by the equation:



There is literature in abundance on the catalysts and the process for the catalytic partial oxidation of gaseous hydrocarbons, in particular methane. Reference is made,

10 for instance, to EP-A-303 438, US-A-5,149,464 and International patent application publication WO 92/11199.

The use of hydrocarbons which are liquid under standard conditions of temperature and pressure (i.e. at 0 °C and 1 atm.) and having at least 6 carbon atoms as feedstocks

15 in catalytic partial oxidation processes has been described in International patent application publication WO 99/19249. For successful operation at commercial scale, the catalytic partial oxidation process must be able to achieve a high conversion of the hydrocarbon

20 feedstock at very high throughputs. In the art, the catalytic partial oxidation of gaseous hydrocarbons at a gas hourly space velocity in the order of 1,000,000 Nl/kg/hr or more is known, for example from US-A-5,648,582. High space velocities have also been

25 described in said WO 99/19249.

Catalytic partial oxidation processes, like the process according to the present invention, are suitably carried out by contacting a feedstock comprising a hydrocarbonaceous fuel and an oxygen-containing gas (oxidant), in amounts giving an oxygen/carbon ratio in the range from 0.3 to 0.8, with the catalyst bed at a temperature in the range of from 750 °C to 1500 °C, at a pressure in the range of from 2 to 100 bara.

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The hydrocarbonaceous fuel suitably comprises hydrocarbons and/or oxygenates which are gaseous under the conditions prevailing at/in the catalyst bed during normal operating conditions. The fuel preferably contains 5 at least compounds that are liquid under standard conditions of temperature and pressure.

Particularly suitable fuels comprise light hydrocarbons, such as methane, natural gas, and associated gas or hydrocarbons being gaseous when 10 contacting the catalyst during operation, but being liquid under standard conditions of temperature and pressure. The latter hydrocarbons typically have an average carbon number of at least 6 and contain up to 25 carbon atoms in their molecules, for example 15 hydrocarbons boiling in the range of from 50 °C to 500 °C, preferably in the range from 60 °C to 350 °C. The term "light hydrocarbons" is a reference to hydrocarbons having from 1 to 5 carbon atoms. The process is particularly suitable for the catalytic partial oxidation 20 of naphtha boiling up to 150 °C, kerosene feedstocks boiling between 150 °C and 200 °C or synthetic gas oil feedstocks boiling between 200 °C and 500 °C, in particular between 200 °C and 300 °C.

It is, of course, possible to have fuel present in 25 the feedstock which gaseous under standard conditions of temperature and pressure, together with material which is liquid under standard conditions of temperature and pressure.

The process according to the present invention can 30 also be carried out when the fuel contains oxygenates (being gaseous and/or liquid under standard conditions of temperature and pressure). Oxygenates which can be used as (part of the fuel) are defined as molecules containing apart from carbon and hydrogen atoms at least one oxygen

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atom which is linked to either one or two carbon atoms or to a carbon atom and a hydrogen atom, such as alcohols, ethers, acids and esters. Examples of suitable oxygenates comprise methanol, ethanol and dimethyl ether. Also 5 mixtures of hydrocarbons and oxygenates can be used as fuel, which may also contain carbon dioxide. When hydrogen is to be produced which is to serve as input to a fuel cell, it is preferred to use a fuel not containing substantial amounts of oxygenates, and, if possible in 10 the substantial absence of oxygenates. Kerosene and synthetic gas oils are preferred feedstocks for the process according to the present invention.

The oxygen-containing gas may be air, substantially pure oxygen, or oxygen-enriched air. The feedstock 15 comprises the fuel (hydrocarbonaceous material and/or oxygenate) and the oxygen-containing gas in amounts sufficient to give an oxygen/carbon ratio in the range from 0.3 to 0.8, preferably of from 0.45 to 0.75, more preferably in the range of from 0.45 to 0.65. References 20 to the oxygen/carbon ratio refer to the ratio of oxygen in the form of molecules (O_2) to carbon present in the fuel.

Although not essential for the proper operation of the regulating system, the feedstock may comprise steam 25 and/or carbon dioxide. If steam is present in the feedstock, the steam/carbon ratio (that is the ratio of molecules of steam (H_2O) to carbon in the fuel) is preferably in the range of from above 0.0 to 3.0, more preferably of from above 0.0 to 2.0. The presence of 30 steam may accelerate the production of hydrogen.

The catalytic partial oxidation is preferably operated at a temperature in the range of from 750 °C to 1500 °C, more preferably in the range of from 800 °C to 1350 °C. Temperatures in the range of from 850 °C to

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1300 °C are particularly suitable. Reference herein to temperature is to the temperature at or in the top (i.e. the upstream side) layer of the catalyst bed, facing the feedstock entering the reactor.

5 The catalytic partial oxidation process is typically operated at a pressure in the range of from 1 to 100 bara, preferably in the range of from 1 to 50 bara, more preferably of from 1 to 10 bara. It will be appreciated that for stationary applications preference 10 will be given to the use of pressures at the lower end of the range.

15 In the catalytic partial oxidation process in accordance with the present invention, the feedstock throughput and thus the gas hourly space velocity (expressed as normal litres of gas per kilogram of catalyst per hour; normal litres are litres at STP, i.e. 0 °C and 1 atm.) can be varied (and the composition of the feedstock, i.e. the oxygen/carbon ratio) will be varied if depending on the hydrogen demand as triggered 20 by the electricity needed by the motorist). At maximum feed throughput, i.e. typically at 0% coverage of the upstream surface of the catalyst bed, the feedstock is preferably provided at gas hourly space velocities which are in the range of from 50,000 to 100,000,000 Nl/kg/hr, 25 more preferably in the range from 100,000 to 10,000,000 Nl/kg/hr, even more preferably in the range of from 200,000 to 3,000,000 Nl/kg/hr. Maximum space velocities in the range of from 500,000 to 1,500,000 Nl/kg/hr are particularly suitable.

30 During the catalytic partial oxidation process, the fuel and the oxidant are preferably contacted with the catalyst under substantially adiabatic conditions. For the purposes of this specification, the expression "substantially adiabatic" is a reference to reaction 35 conditions in which substantially all heat loss and

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radiation from the reaction zone is prevented, with the exception of heat leaving the reactor in the gaseous effluent stream.

5 Catalyst compositions suitable for use in catalytic partial oxidation are known in the art. Such catalysts generally comprise, as active component, a metal selected from Group VIII of the Periodic Table of the Elements. References in this specification to the Periodic Table of the Elements are to the CAS version, as published in the

10 CRC Handbook of Chemistry and Physics, 68th Edition.

Catalysts comprising, as the catalytically active component, a metal selected from rhodium, iridium, palladium and platinum are preferred. Catalysts comprising rhodium or iridium are most preferred.

15 The catalytically active metal is most suitably supported on a carrier, such as refractory oxide particles, monolith structures, or metallic arrangements such as metal gauzes. Suitable carrier materials are well known in the art and include refractory oxides, such as

20 silica, alumina, titania, zirconia and mixtures thereof. Mixed refractory oxides, that is refractory oxides comprising at least two cations may also be employed as carrier materials for the catalyst.

25 The catalyst may comprise the catalytically active metal in any suitable amount to achieve the required level of activity. Typically, the catalyst comprises the active metal in an amount in the range of from 0.01 to 20% by weight, preferably from 0.02 to 10% by weight, more preferably from 0.1 to 7.5 % by weight.

30 The process according to the present invention is suitably used to prepare hydrogen or a mixture of hydrogen and other gases from any hydrocarbonaceous feedstock. Preferably, the process according to the present invention is directed at producing hydrogen from

35 fuel, in particular at the on-board production of

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hydrogen in fuel cell powered transport means, such as automotive vehicles and crafts.

5 Accordingly, the present invention also relates to transport means provided with a catalytic partial oxidation system provided with oxygen/carbon regulating means.

10 With respect to the regulating aspects connected with the process according to the present invention, it will be clear that the essential part of regulating the oxygen/carbon ratio comprises the capability of having a quick response device to measure the temperature upstream of the catalyst in the reactor. In practice, this can be carried out conveniently by placing a quick response device at a suitably distance upstream of the catalyst bed, e.g. by placing it before a window in the reactor which allows uninterrupted monitoring of the catalyst surface. "Upstream" in the context of this specification is meant in relation to the temperature at the catalyst upstream surface or above the catalyst upstream surface (in the area of feedstock about to enter the catalyst) to the extent that it is still representative for the temperature at the catalyst upstream surface.

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20 The essence of the process according to the present invention resides in the capability of quickly and reliably measuring the temperature (upstream) of the catalyst surface and comparing it with a calibrated temperature profile. The calibration can in principle be carried out in situ (i.e. under (semi)-operational conditions) or ex situ (e.g. by measuring a standard temperature profile in a fixed system which will render a (set of) reference point(s) to be used in control unit 12). Ex situ temperature profiles suitable for calibration can be produced at the manufacturing facility for the hydrogen producing device or can be (re)-checked 25 when the device is brought in for maintenance purposes.

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Preference is given to in situ regulating systems, in particular to systems in which calibration is achieved by measuring the temperature needed for calibration input at a point downstream of the catalyst bed. A suitable and preferred place for measuring the temperature needed for calibrating purposes is at the bottom of the catalyst bed. One of the advantages connected with in situ measurement of the temperature needed for calibration purposes is that faults due to system aberrations can be suppressed as far as possible. "Bottom" in the context of this specification is meant in relation to the temperature at the bottom of the catalyst bed or below the catalyst (in the area where effluent appears coming out of the catalyst bed) to the extent that it is still representative for the temperature at the bottom of the catalyst bed.

Quick response signals can be processed in essence either in a so-called re-active mode (when using quick response actuators directly coupled to the input signal) or in so-called pro-active mode (using the same or similar quick response actuators but coupled indirectly to the input signal via an optimised dynamic model).

Pro-active feedback is based upon a dynamic model of the system under review which can be fine-tuned by a minimum number of parameters to be optimised during an acceleration of the motor vehicle operating on electricity produced from hydrogen. An advantage of a pro-active feedback system is that it can operate rather simply as only the AC component of the signal will be relevant thereby allowing slow drifts in the system (which may occur in time) to be ignored.

A suitable means for quick and reliable temperature measurement (which can not be provided for by common thermocouples because of their slow response) comprises a two-colour photo detecting device. Such apparatus

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operates on the basis of measuring the colour of a surface (in this case the colour of the surface of the catalyst bed facing the feedstock) using an optical fibre. Best results will be obtained when instead of the absolute light intensity the ratio between two light intensities is measured. This type of measurement is advantageous in that it reduces or even eliminates unwanted influences such as the position of a diaphragm when used to shield part of the surface of the catalyst or the presence of some local "dead spots" on the catalyst surface or the occurrence of achromatic fouling.

It has been found that good results can be obtained when the colour measurements are carried out using a pyrometer operable in the range of from 700 to 1000 nm (near infrared) as this is the area of highest sensitivity for changes in colour for silicon-type photo-detection. Preference is given to measurements performed in the 700 to 950 nm range. It will be clear that during normal operation of a catalytic partial oxidation process the surface of the catalyst is glowing and that changes in process conditions will influence the intensity and the wavelength of the colour(s) of the glowing catalyst surface. Pyrometers are suitably made of glass or quartz, e.g. in the form of a glass rod or a glass fibre and may contain a silicon chip or other recording device capable of receiving and transmitting data. The time involved in measuring the temperature on the basis of certain wavelengths is in the order of 1 - 100 milliseconds, preferably in the range between 5 and 30 milliseconds. This is sufficient to allow a quick measurement to start adjustment, if necessary.

Good results can be obtained by using dual wavelength pyrometers which measure light intensities for two (pre-set) wavelengths rather than over the total bandwidth, for instance by using a splitter system or, alter-

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natively, a prism. The light intensities measured are converted via amplifiers and A-D current converters into a signal which can be processed in the control unit. Dual-wavelength pyrometers are commercially available from number of companies and can be operated within the temperature ranges and wavelengths envisaged for the process according to the present invention.

The second part of the temperature comparison in order to decide on adjustment of the oxygen/carbon ratio comprises measuring the temperature at a point downstream of the catalyst system. "Downstream" in the context of the calibration in accordance with the present invention has a similar meaning as "bottom" of the catalyst system as defined hereinbefore. Since the "downstream temperature" is less susceptible to rapid changes there is no need to provide a quick response temperature system in this part of the measurement chain. Therefore, conventional thermocouples can be used to collect the data needed. It is, of course, possible to apply a quick response device such as the device used at the upstream part of the catalyst bed but that will add to the costs of the regulating system and is not needed as far as accuracy or speed of the measurement are concerned, in particular when calibration of the system has been carried out separately.

In practice, one will normally observe an offset in temperature between the top of the catalyst bed and the bottom part thereof. The magnitude of the offset will depend inter alia on the construction of the catalytic system applied and, to some extent on the amount of steam used in the process. This does not play a major role in the proper operation of the regulating system. Those skilled in the art will be familiar with such effects.

Calibration of the measurement system when operating at in situ mode is suitably carried out by recording the

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relationship between colour and temperature. This can be achieved with model systems. It is also possible to (re)-calibrate the relationship during (quasi)-stationary periods during operation. For instance, when a vehicle is operated in a constant mode (i.e. demanding the same amount of electricity, and hence, hydrogen, over a prolonged period of time which is at least longer than the slowest response time) (re)-calibration can be carried out conveniently. If the conditions dictate that (re)-calibrations have to be carried out rather frequently, it will be advantageous to also use a quick response device downstream of the catalyst system in order not to be hampered by any time delay which may be caused by the thermal capacity of the catalyst bed being monitored. Calibration of the measurement system can also be carried out ex situ.

(Quasi)-stationary conditions in accordance with the process according to the present invention are conditions during which the temperature variation downstream of the catalyst bed is less than 20 °C/minute.

It will be clear that during start-up of the process for catalytic partial oxidation the catalyst system does not (yet) glow and therefore not capable of emitting light having the wavelengths on which the monitoring system is based. Therefore, an estimate of the amount of oxidant needed will have to be made which, when supplied together with fuel, will cause the catalyst bed to start glowing. At a certain signal/noise ratio proper measurement sequences will be in place.

The data fed to the control unit, which can be any data processor capable of receiving data signals within the time-frames applicable in measuring the temperature upstream and downstream of the catalyst bed, are processed within the unit and will trigger data transfer to the actuator when the deviation from the pre-set value

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(or pre-set value range as the case may be) is high enough. For instance, when the motorist wants to accelerate more electricity is needed and hence more hydrogen has to be fed to the fuel cell which requires 5 producing more hydrogen in the same time which means that more fuel has to be provided in the feedstock to the reactor. If the fuel change is not proportional to the oxidant flow rate, a change in the colour of the glowing surface of the catalyst will occur which has to be 10 corrected. If (quasi)-stationary conditions have been reached (independent of their absolute level) flow adjustment will not be necessary and no data signals (or no different data signals) will be transmitted to the actuator.

15 The adjustment of the amount of fuel to be present, together with oxygen, in the feedstock to the reactor is carried out by means of an actuator present in the feed line 1 to the reactor prior to the mixing of oxidant and fuel. In principle it is possible to position the 20 actuator in the oxidant line 2 (or even in both) but it is preferred to position the actuator in the fuel line since the oxidant is normally supplied by a compressor which has a rather slow response time. Therefore, in 25 practice, the flow of oxidant is set in relation to the amount of hydrogen expected to be necessary and the fuel flow set to obtain the required oxygen/carbon ratio.

30 The amount of fuel in line 1 which is to be mixed with the oxidant (for instance, oxygen or air) in line 2 is suitably regulated by means of atomisers or vaporisers. Those skilled in the art know which dosing system to choose and how to operate it. Systems which can be suitably applied are commercially available from companies like Bosch and Mitsubishi. The systems operate by pulsed injection of fuel in a regime of continuous

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conversion (partial oxidation) with changing demands on the level of conversion.

Preferably, fuel passing through line 1 is added to the air stream passing through line 2 by using a pulsed liquid injection system. Such system is operated by setting the frequency of the pulse and the opening time of the atomiser. It operates in slave- mode, triggered by a signal from control unit 12. It is capable of reacting quickly on changes in the temperature of the upstream 5 surface of the catalyst bed as well as on changes in the oxidant throughput if that were to be required). It has been found that fluctuations in the oxygen/carbon ratio become smaller (and, hence, the performance in regulating more sophisticated) when use is made of smaller pulses 10 and/or when the dead time between subsequent pulses is minimised. It has been found that the use of pulsed injection systems allows control of the fuel-to-air ratio at high precision for high turn down ratios in the range of from 1:100. Moreover, such systems are capable of 15 maintaining adequate droplet size distribution even when atomisation is carried out under low throughput conditions. The use of short, strong injections compensates for the lack of turbulence at relatively low fuel velocities. Such pulsed liquid injection systems 20 normally include mixing chambers having a residence time of several times the pulse interval time. Throughput variations can be achieved by varying pulse duration and 25 pulse frequency.

It is also possible to add other ingredients to the 30 fuel and or oxidant such as water. Water is preferably added in constant ratio with respect to fuel consumption. Adjustments can be made depending on anticipated changes in the revolutions per time unit of the compressor feeding the oxidant (normally air) through line 2.

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Although the process according to the present invention is particularly suitable for regulating the oxygen/carbon ratio in catalytic (partial) oxidation processes, it is in essence suitable for any system which 5 is confronted with large changes in fuel compositions and demands. For instance, when using off-shore gas as feedstock when the stoichiometry of system has to be maintained or fluctuated within pre-determined ranges.



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TS 0793 EPC

C L A I M S

1. Process for regulating the oxygen/carbon ratio in the feedstock for a catalytic oxidation process in which a feedstock comprising a mixture of fuel and oxidant is passed through a catalyst bed in a reactor and hydrogen-containing reactor effluent is collected, in which process the oxygen/carbon ratio in the feedstock is regulated by

- 5 a) measuring the temperature upstream of the catalyst by means of a quick response device,
- 10 b) feeding the measured data of the temperature measurement to a control unit and comparing them with a calibrated temperature profile, and adjusting the amount of fuel in the feed to substantially maintain the oxygen/carbon ratio under non-steady hydrogen production.

15 2. Process according to claim 1, in which calibration is carried out by measuring the temperature downstream of the catalyst bed.

20 3. Process according to claim 2, in which the temperature is measured at the bottom of the catalyst bed.

25 4. Process according to one or more of the preceding claims, in which the catalytic oxidation process is a catalytic partial oxidation process.

5. Process according to one or more of the preceding claims, in which use is made of a molecular oxygen-containing oxidant.

30 6. Process according to one or more of the preceding claims, in which use is made of an oxygen/carbon ratio in the range of from 0.3 to 0.8, preferably in the range of

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from 0.45 to 0.75, most preferably in the range of from 0.45 to 0.65.

5 7. Process according to one or more of the preceding claims, in which use is made of a gas velocity in the range between 100,000 and 20,000,000 Nl/kg/hr.

8. Process according to one or more of the preceding claims, in which use is made of a catalyst comprising one or more of Group VIII noble metals on a carrier.

10 9. Process according to claim 8, in which use is made of Rh as metal component on an amorphous carrier.

10. Process according to claim 8 or 9, in which use is made of metallic arrangements, preferably of metal gauzes as catalyst carriers.

15 11. Process according to on or more of the preceding claims, which is carried out at a temperature in the range between 700 °C and 1500 °C.

12. Process according to one or more of the preceding claims, in which use is made of a pro-active or of a re-active quick response device.

20 13. Process according to claim 12, in which use is made of an optical pyrometer to measure the light intensities occurring at the surface of the catalyst bed facing the feedstock.

25 14. Process according to claim 12 or 13, in which the light intensities of two wavelengths are measured at or above the surface of the catalyst bed.

15. Process according to claim 13 or 14, in which light intensities are measured in the range between 700 and 1000 nm.

30 16. Process according to one or more of claims 13-15, in which the temperature at or below the bottom part of the catalyst bed is measured by a thermocouple or an optical pyrometer.

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17. Process according to one or more of claims 13-16, in which the measurement is carried out within a time frame of from 1 to 100 milliseconds.

5 18. Process according to one or more of claims 13-17, in which calibration of the temperature monitoring system is carried out under (quasi) - stationary conditions.

19. Process according to one or more of the preceding claims, in which use is made of a rapid response actuator to adjust the amount of fuel in the feed.

10 20. Process according to claim 19, in which use is made of a pulsed liquid injection system as rapid response actuator.

21. Process according to claim 20, in which use is made of a pulsed liquid injection system under conditions comprising turn down ratios in the range of from 1:100.

15 22. Transport means provided with a catalytic partial oxidation system provided with oxygen/carbon regulating means operating according to one or more of the preceding claims.

C45/TS0793PD



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TS 0793 EPC

A B S T R A C T

FLOW CONTROL SYSTEM

Process for regulating the oxygen/carbon ration in the feedstock for a catalytic oxidation process in which a feedstock comprising a mixture of fuel and oxidant is passed through a catalyst bed in a reactor and a hydrogen-containing reactor effluent is collected, in which process the oxygen/carbon ratio in the feedstock is regulated by

- a) measuring the temperature upstream of the catalyst by means of a quick response device,
- b) feeding the measured data of the temperature measurement to a control unit and comparing them with a calibrated temperature profile, and adjusting the amount of fuel in the feed to substantially maintain the oxygen/carbon ratio under non-steady hydrogen production.

The process is particularly suitable for adjusting the oxygen/carbon ratio in the feedstock to produce hydrogen to be fed to a fuel cell to produce electricity, especially when varying amounts of electricity are demanded (such as by motorists driving hydrogen-operated vehicles or in domestic applications).

C45/TS0793PD



IS 0793 EPL

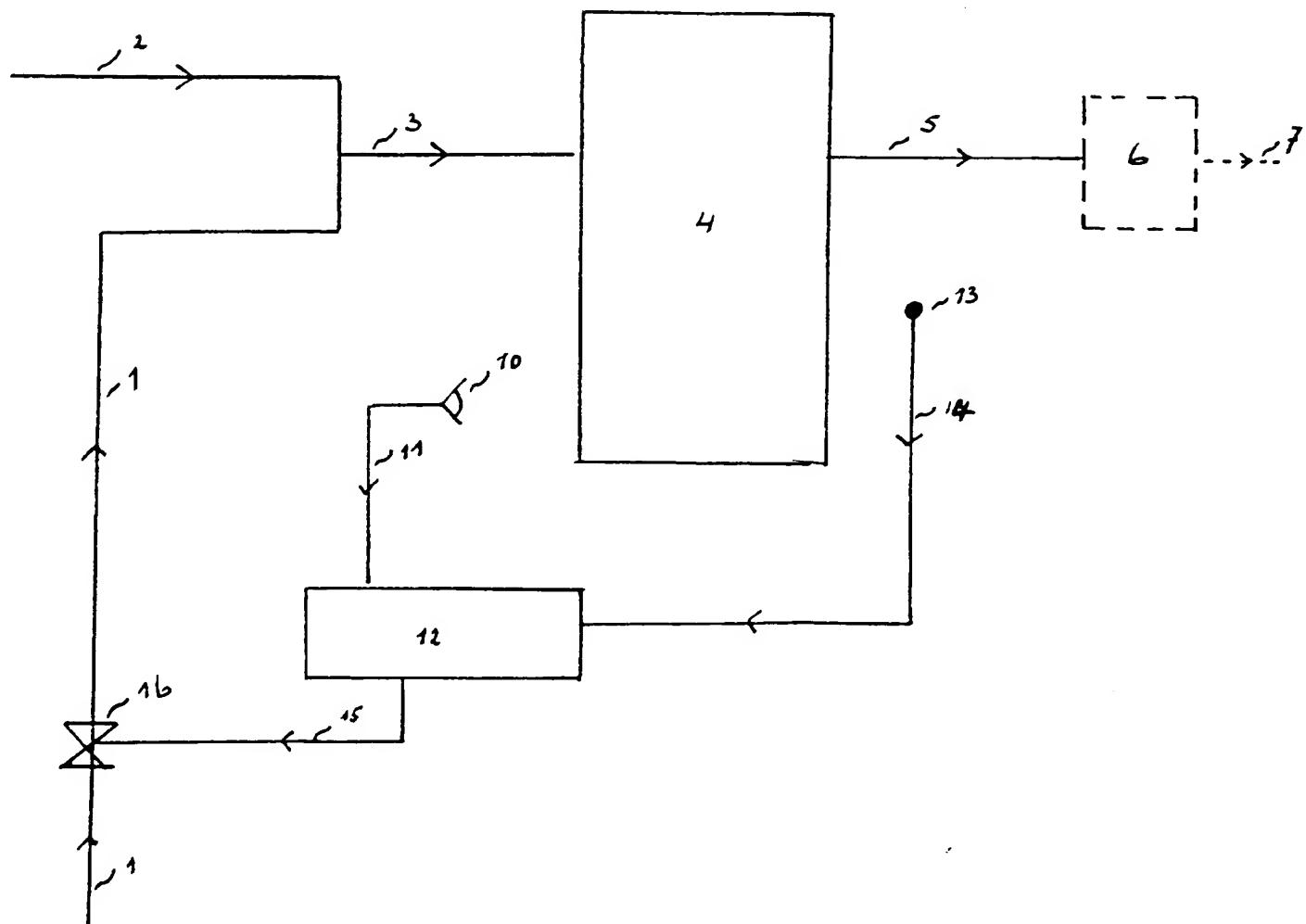


Figure 1

